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<b>(54) Title:</b> POLYPROPYLENE AND POLYESTER BLENDS CONTAINING A GRAFT-MODIFIED POLYOLEFIN ELASTOMER <b>(57) Abstract</b> <p>Disclosed are polymer blend compositions comprising polypropylene, a thermoplastic polyester, a graft-modified polyolefin elastomer, and optionally an impact modifier and methods of preparation of such compositions.</p>		

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POLYPROPYLENE AND POLYESTER BLENDS CONTAINING A GRAFT-MODIFIED POLYOLEFIN ELASTOMER

This invention relates to compositions containing a polypropylene, a thermoplastic polyester, a graft-modified polyolefin elastomer, and optionally an impact modifier and methods of preparation of such compositions. This invention relates particularly to a blend composition having improved processability and when molded, having improved impact strength.

Polypropylene, especially highly isotactic polypropylene, has been used in many applications in the form of molded articles, film, sheet, etc., because it is excellent in molding processability, toughness, moisture resistance, gasoline resistance, chemical resistance, has a low specific gravity, and is inexpensive. However, polypropylene is poor or inadequate in heat resistance, stiffness, impact resistance, and scratch resistance. These deficiencies are obstacles to opening up new applications for polypropylene.

On the other hand, thermoplastic polyesters such as polyethylene terephthalate are widely used as engineering thermoplastics in the fields of automobile parts, electrical components and electronic parts, because such polymers have high heat resistance, stiffness, strength, solvent resistance, and oil resistance. It would, however, be desirable to further improve the molding processability, toughness, notched impact resistance and chemical resistance of these plastics. In addition, polyesters are disadvantaged in that these plastics have higher specific gravity and are more expensive than polypropylenes.

From such a view point, it would seem a useful approach to blend polypropylene and a thermoplastic polyester in order to obtain a thermoplastic resin having the characteristics of both of these polymers. However, physical blending of these polymers has proven to be difficult in practice. Since polypropylene and thermoplastic polyesters are naturally incompatible, merely mixing these polymers in the melt to form a suitable blend has not proven possible. These immiscible plastics exhibit poor adhesion along domain interfaces with resultant weakness in the solid resin, evidenced as gross phase separation. The mechanical properties, in particular impact resistance, tensile elongation, and tensile strength of a molded product made of a mixture of polypropylene and thermoplastic polyester often have values lower than those expected by averaging of the physical properties of the polypropylene and thermoplastic polyester. When injection molded, the resulting products show an extreme nonuniformity and an unacceptable appearance owing to the formation of

flow marks, and cannot be used in practice in the manufacture of articles for use as automobile parts, electric components, and electronic parts.

It is previously known in the art to use interfacial agents and/or impact modifiers to produce blends of immiscible polymers with a desirable balance of properties. Interfacial agents provide adhesion between the principal polymer phases, improving stress transfer, and are necessary to reduce interfacial tension during processing that can lead to gross phase separation. Thus, interfacial agents play an important role in determining the ultimate morphology of the blend. Reduction of interfacial tension and impact modification can be provided by a single compound or by distinct compounds.

Methods of improving impact strength in individual polymers, such as polypropylene, include the use of impact modifiers having a low glass transition temperature ( $T_g$ ). Generally, to significantly increase the impact strength of a thermoplastic, it is necessary to blend in an impact modifier that forms finely dispersed rubber particles within the polymer matrix. These rubber particles improve energy dissipation in the thermoplastic while simultaneously limiting the growth of cracks. To achieve the required morphology for effective toughening, the impact modifier should be compatible with the thermoplastic to be toughened.

One method for improving impact properties in polypropylene is using polyolefin elastomers (POEs). These polymers are compatible with polypropylene and have a low glass transition temperature less than 25°C, preferably less than 0°C. Examples of these types of polymers include copolymers of alpha-olefins, such as ethylene and propylene, ethylene and 1-butene, ethylene and 1-hexene or ethylene and 1-octene copolymers, and terpolymers of ethylene, propylene and a diene comonomer such as hexadiene or ethylidene norbornene.

Typically, impact modifiers useful for improving the impact resistance of polymers having a similar structure are often less useful for modifying polymers dissimilar in structure. For example, polyolefin elastomers are useful in improving the impact resistance of polypropylene, but are less useful for improving the impact resistance of thermoplastic polyester. However, it is well known that grafting functional groups to polymers can enhance their interaction with dissimilar polymers, this is sometimes referred to as compatibilization. These interactions can include chemical bonding, for example, cross linking, hydrogen bonding and dipole-dipole interaction. Typically, in order to obtain an advantageous degree of functional moieties on the base polymer a certain amount of residual unsaturation must be present.

Maleic anhydride, for example, has been proposed as a compatibilizing group for a variety of polymers and plastic blends, see *Plastics Technology*, February, 1989, pages 67-75; Albee et al., *Plastics Compounding*, September/October 1990, pages 32-41; Hughes et al., US-A-5,346,963, issued September 13, 1994, substantially  
5 linear ethylene polymers grafted with maleic anhydride; Hughes et al., US-A-5,705,565, issued January 6, 1998, to increase impact resistant of selected thermoplastic blends by addition of a minor amount of substantially linear ethylene polymer grafted with maleic anhydride; Tekkanat et al., US-A-5,280,066, issued January 18, 1994, to increase impact resistance of polyolefin blends by addition of a  
10 minor amount of a maleic anhydride grafted hydrogenated block copolymer of styrene and butadiene (SBR); Fujita et al., US-A-5,444,119, issued August 22, 1995, to increase impact resistance of polypropylene and polyester blends with a polypropylene and polyester copolymer and a maleic anhydride grafted polypropylene; and Henman et al., US-A-4,054,549, issued October 18, 1977, to  
15 improve adhesion between polypropylene and polyester with a blend of at least one boron, phosphorous or sulfur containing acid and a maleic anhydride grafted polypropylene.

While some properties, such as impact resistance, have undoubtedly been improved in the blends of the above-cited references, other bulk properties of the  
20 resulting blend have suffered. The present invention addresses this problem.

Accordingly, the present invention is directed to a polymer blend composition comprising (a) polypropylene, (b) a thermoplastic polyester, (c) a polyolefin elastomer grafted with an unsaturated organic compound containing, prior to grafting, at least one site of ethylenic unsaturation and at least one carbonyl group, preferably maleic  
25 anhydride, and optionally (d) an impact modifier. Said composition possess a good balance of good processability, good thermal and physical properties, good solvent resistance, and especially, improved impact resistance.

In a further embodiment, the invention also involves a method of preparing the forgoing polymer blend composition comprising combining (a) polypropylene, (b) a  
30 thermoplastic polyester, (c) a graft-modified polyolefin elastomer, and optionally (d) an impact modifier.

In yet a further embodiment, the invention involves a method of molding a polymer blend composition whereby (a) polypropylene which has been admixed with at least (b) a thermoplastic polyester, (c) a graft-modified polyolefin elastomer, and  
35 optionally (d) an impact modifier is molded.

Component (a) in the polymer blend compositions of this invention is a polypropylene. The polypropylene suitable for use in this invention is well known in the literature and can be prepared by known techniques. In general, the polypropylene is in the isotactic form of homopolymer polypropylene, although other forms of polypropylene can also be used (for example, syndiotactic or atactic). Polypropylene impact copolymers (for example, those wherein a secondary copolymerization step reacting ethylene with the propylene is employed), however, can also be used in the polymer blend compositions disclosed herein. A complete discussion of various polypropylene polymers is contained in *Modern Plastics Encyclopedia/89*, mid October 1988 Issue, Volume 65, Number 11, pp. 86-92. The molecular weight of the polypropylene for use in the present invention is conveniently indicated using a melt flow measurement, sometimes referred to as melt flow rate (MFR) or melt index (MI), according to ASTM D 1238, Condition 230°C/2.16 kilogram (kg). Melt flow rate is inversely proportional to the molecular weight of the polymer. Thus, the higher the molecular weight, the lower the melt flow rate, although the relationship is not linear. The melt flow rate for the polypropylene useful herein is generally greater than 0.1 grams/10 minutes (g/10 min), preferably greater than 0.5 g/10 min, more preferably greater than 1 g/10 min, and even more preferably greater than 10 g/10 min. The melt flow rate for the polypropylene useful herein is generally less than 100 g/10 min, preferably less than 75 g/10 min, more preferably less than 60 g/10 min, and more preferably less than 50 g/10 min.

The polypropylene is employed in the polymer blend compositions of the present invention in amounts sufficient to provide the desired balance of processability and impact resistance. In general, the polypropylene is employed in amounts of at least 5 percent by weight, preferably at least 10 percent by weight, more preferably at least 20 percent by weight, even more preferably at least 30 percent by weight, and most preferably at least 40 percent by weight based on the weight of the polymer blend composition. In general, the polypropylene is used in amounts less than or equal to 95 percent by weight, preferably less than or equal to 90 percent by weight, more preferably less than or equal to 70 percent by weight, even more preferably less than or equal to 50 percent by weight, and most preferably less than or equal to 45 percent by weight based on the weight of the polymer blend composition.

The thermoplastic polyesters, component (b), which can be used in this invention are known and are commercially available, and may be made by a variety of methods. A complete discussion of various polyester polymers is contained in

*Encyclopedia of Polymer Science and Engineering*, 1988, Volume 12, pp. 1-312, of particular interest is the section on thermoplastic polyesters found on pp. 217-256. Examples of such thermoplastic polyesters which are suitable as (b) include poly(alkylene alkanedicarboxylate), a poly(alkylene phenylenedicarboxylate), a poly(phenyl alkanedicarboxylate), or a poly(phenylene phenylenedicarboxylate) and are therefore appropriate for use herein. Methods and materials useful for the production of thermoplastic polyesters are discussed in greater detail in Whinfield, US-A-2,465,319, Pengilly, US-A-3,047,539 and Russell, US-A-3,756,986. Aromatic thermoplastic polyesters such as the poly(alkylene phenylenedicarboxylates), which include polyethylene terephthalate, polypropylene terephthalate, and polybutylene terephthalate, or mixtures or copolymers thereof, are particularly useful in this invention. These aromatic thermoplastic polyesters preferably have an intrinsic viscosity between 0.35 and 1.2, more preferably 0.35 and 1.1, and are more easily processed than aromatic thermoplastic polyesters with higher intrinsic viscosities.

The thermoplastic polyester is employed in the polymer blend compositions of the present invention in amounts sufficient to provide the desired balance of processability and impact resistance. In general, the thermoplastic polyester is employed in amounts of at least 5 percent by weight, preferably at least 10 percent by weight, more preferably at least 20 percent by weight, even more preferably at least 30 percent by weight, and most preferably at least 40 percent by weight based on the weight of the polymer blend composition. In general, the thermoplastic polyester is used in amounts less than or equal to 95 percent by weight, preferably less than or equal to 90 percent by weight, more preferably less than or equal to 70 percent by weight, even more preferably less than or equal to 50 percent by weight, and most preferably less than or equal to 45 percent by weight based on the weight of the polymer blend composition.

The third component (c) in the polymer blend composition is a graft-modified polyolefin elastomer. Suitable polyolefin elastomers comprises one or more C<sub>2</sub> to C<sub>20</sub> alpha-olefins in polymerized form, having a glass transition temperature (T<sub>g</sub>) less than 25°C, preferably less than 0°C. T<sub>g</sub> is the temperature or temperature range at which a polymeric material shows an abrupt change in its physical properties, including, for example, mechanical strength. T<sub>g</sub> can be determined by differential scanning calorimetry. Examples of the types of polymers from which the present polyolefin elastomers are selected include copolymers of alpha-olefins, such as ethylene and propylene, ethylene and 1-butene, ethylene and 1-hexene or ethylene and 1-octene

copolymers, and terpolymers of ethylene, propylene and a diene comonomer such as hexadiene or ethylidene norbornene.

A preferred polyolefin elastomer for use herein is one or more substantially linear ethylene polymer or a linear ethylene polymer (S/LEP). Both substantially  
5 linear ethylene polymers and linear ethylene polymers are known. Substantially linear ethylene polymers and their method of preparation are fully described in US-A-5,272,236 and US-A-5,278,272. Linear ethylene polymers and their method of preparation are fully disclosed in US-A-3,645,992; US-A-4,937,299; US-A-4,701,432; US-A-4,937,301; US-A-4,935,397; US-A-5,055,438; EP-A-129,368; EP-A-260,999;  
10 and WO 90/07526.

As used here, "a linear ethylene polymer" means a copolymer of ethylene and one or more alpha-olefin comonomers having a linear backbone (that is, no cross linking), no long-chain branching, a narrow molecular weight distribution and a narrow composition distribution. Further, as used here, "a substantially linear ethylene  
15 polymer" means a copolymer of ethylene and of one or more alpha-olefin comonomers having a linear backbone, a specific and limited amount of long-chain branching, a narrow molecular weight distribution and a narrow composition distribution.

Short-chain branches in a linear copolymer arise from the pendent alkyl group  
20 resulting upon polymerization of intentionally added C<sub>3</sub> to C<sub>20</sub> alpha-olefin comonomers. Narrow composition distribution is also sometimes referred to as homogeneous short-chain branching. Narrow composition distribution and homogeneous short-chain branching refer to the fact that the alpha-olefin comonomer is randomly distributed within a given copolymer of ethylene and an alpha-olefin  
25 comonomer and virtually all of the copolymer molecules have the same ethylene to comonomer ratio. The narrowness of the composition distribution is indicated by the value of the Composition Distribution Branch Index (CDBI) or sometimes referred to as Short Chain Branch Distribution Index. CDBI is defined as the weight percent of the polymer molecules having a comonomer content within 50 percent of the median  
30 molar comonomer content. The CDBI is readily calculated, for example, by employing temperature rising elution fractionation, as described by Wild in the Journal of Polymer Science, Polymer Physics Edition, Volume 20, page 441 (1982), or US-A-4,798,081. The CDBI for the substantially linear ethylene polymers and the linear ethylene polymers in the present invention is greater than 30 percent,  
35 preferably greater than 50 percent, and more preferably greater than 90 percent.



Long-chain branches in substantially linear ethylene polymers are polymer branches other than short chain branches. Typically, long chain branches are formed by in situ generation of an oligomeric alpha-olefin via  $\beta$ -hydride elimination in a growing polymer chain. The resulting species is a relatively high molecular weight vinyl terminated hydrocarbon which upon polymerization yields the large pendent alkyl group. Long-chain branching may be further defined as hydrocarbon branches to a polymer backbone having a chain length greater than  $n-2$  ( $n-2$ ) carbons, where  $n$  is the number of carbons of the largest alpha-olefin comonomer intentionally added to the reactor. Preferred long-chain branches in copolymers of ethylene and one or more  $C_3$  to  $C_{20}$  alpha-olefin comonomers have at least from 20 carbons up to more preferably the number of carbons in the polymer backbone. Long-chain branching may be distinguished using  $^{13}C$  nuclear magnetic resonance spectroscopy alone, or with gel permeation chromatography-laser light scattering (GPC-LALS) or a similar analytical technique. Substantially linear ethylene polymers contain at least 0.01 long-chain branches/1000 carbons and preferably at least 0.05 long-chain branches/1000 carbons. In general, substantially linear ethylene polymers contain less than or equal to 3 long-chain branches/1000 carbons and preferably less than or equal to 1 long-chain branch/1000 carbons.

Preferred substantially linear ethylene polymers are prepared by using metallocene based catalysts capable of readily polymerizing high molecular weight alpha-olefin copolymers under the process conditions. As used here, copolymer means a polymer of two or more intentionally added comonomers, for example, such as might be prepared by polymerizing ethylene with at least one other  $C_3$  to  $C_{20}$  comonomer. Linear ethylene polymers may be prepared in a similar manner using metallocene or vanadium based catalyst under conditions that do not permit polymerization of monomers other than those intentionally added to the reactor. Other basic characteristics of substantially linear ethylene polymers or linear ethylene polymers include a low residuals content (that is, a low concentration therein of the catalyst used to prepare the polymer, unreacted comonomers and low molecular weight oligomers made during the course of the polymerization), and a controlled molecular architecture which provides good processability even though the molecular weight distribution is narrow relative to conventional olefin polymers.

Preferably the substantially linear ethylene polymers or the linear ethylene polymers comprise between 50 to 95 weight percent ethylene and from 5 to 50, and preferably from 10 to 25 weight percent of at least one  $\alpha$ -olefin comonomer. The comonomer content in the substantially linear ethylene polymers or the linear

ethylene polymers is generally calculated based on the amount added to the reactor and as can be measured using infrared spectroscopy according to ASTM D 2238, Method B. Typically, the substantially linear ethylene polymers or the linear ethylene polymers are copolymers of ethylene and one or more C<sub>3</sub> to C<sub>20</sub> alpha-olefins, preferably copolymers of ethylene and one or more C<sub>3</sub> to C<sub>10</sub>, alpha-olefin comonomers and more preferably copolymers of ethylene and one or more comonomers selected from the group consisting of propylene, 1-butene, 1-hexene, 4-methyl-1-pentane, and 1-octene. Most preferably copolymers are ethylene and 1-octene copolymers.

The density of these substantially linear ethylene polymers or linear ethylene polymers is equal to or greater than 0.850 grams per cubic centimeter (g/cm<sup>3</sup>) and preferably equal to or greater than 0.860 g/cm<sup>3</sup>. Generally, the density of these substantially linear ethylene polymers or linear ethylene polymers is less than or equal to 0.935 g/cm<sup>3</sup> and preferably less than or equal to 0.900 g/cm<sup>3</sup>. Sometimes it is useful to compare the melt flow ratio ( $I_{10}/I_2$ ) as determined according to ASTM D 1238 conditions of 190°C /10.0 kg ( $I_{10}$ ) and 190°C/2.16 kg ( $I_2$ ).  $I_{10}/I_2$  for substantially linear ethylene polymers is greater than or equal to 5.63, preferably from 6.5 to 15, and more preferably from 7 to 10.

The molecular weight distribution ( $M_w/M_n$ ) for substantially linear ethylene polymers is the weight average molecular weight ( $M_w$ ) divided by number average molecular weight ( $M_n$ ).  $M_w$  and  $M_n$  are measured by gel permeation chromatography (GPC). For substantially linear ethylene polymers, the  $I_{10}/I_2$  ratio indicates the degree of long-chain branching, that is, the larger the  $I_{10}/I_2$  ratio, the more long-chain branching exists in the polymer. In preferred substantially linear ethylene polymers  $M_w/M_n$  is related to  $I_{10}/I_2$  by the equation:  $M_w/M_n \leq [(I_{10}/I_2) - 4.63]$ , and is at least 1.5 and preferably at least 2.0. Generally,  $M_w/M_n$  for substantially linear ethylene polymers is less than or equal to 3.5, more preferably less than or equal to 3.0. In a most preferred embodiment, substantially linear ethylene polymers are also characterized by a single differential scanning calorimetry (DSC) melting peak.

The preferred  $I_2$  melt index for these substantially linear ethylene polymers or linear ethylene polymers is from 0.01 g/10 min to 100 g/10 min, and more preferably 0.1 to 10 g/10 min.

The unsaturated organic compound suitable for graft modification of the polyolefin elastomer, prior to grafting, preferably contains at least one site of ethylenic unsaturation and at least one carbonyl group (-C=O). Representative of unsaturated organic compounds that contain at least one carbonyl group are the carboxylic acids,

anhydrides, esters and their salts, both metallic and nonmetallic. Preferably, the organic compound contains ethylenic unsaturation conjugated with a carbonyl group. Representative compounds include maleic, fumaric, acrylic, methacrylic, itaconic, crotonic, methyl crotonic, and cinnamic acid and their anhydride, ester and salt derivatives, if any. Maleic anhydride is the preferred unsaturated organic compound containing at least one site of ethylenic unsaturation and at least one carbonyl group.

The unsaturated organic compound is used in an amount such that, after grafting to the polyolefin elastomer, it constitutes by weight at least 0.01 percent, preferably at least 0.05 percent, more preferably at least 0.1 percent, more preferably at least 0.5 percent, and most preferably at least 1.0 percent based on the weight of grafted polyolefin elastomer. The maximum amount of unsaturated organic compound content can vary to convenience, but typically constitutes by weight less than or equal to 20 percent, preferably less than or equal to 15 percent, more preferably less than or equal to 10 percent, more preferably less than or equal to 5 percent, and most preferably less than or equal to 2 percent based on the weight of the grafted polyolefin elastomer.

The unsaturated organic compound containing at least one carbonyl group can be grafted to the polyolefin elastomer by any known technique, such as those taught in US-A-3,236,917 and US-A-5,194,509. For example, polymer is introduced into a two-roll mixer and mixed at a temperature of 60°C. The unsaturated organic compound is then added along with a free radical initiator, such as, for example, benzoyl peroxide, and the components are mixed at 30°C until the grafting is completed. Alternatively, the reaction temperature is higher, for example, 210 °C to 300°C, and a free radical initiator is not used or is used at a reduced concentration. An alternative and preferred method of grafting is taught in US-A-4,950,541, by using a twin-screw devolatilizing extruder as the mixing apparatus. The polyolefin elastomer and unsaturated organic compound are mixed and reacted within the extruder at temperatures at which the reactants are molten and in the presence of a free radical initiator. Preferably, the unsaturated organic compound is injected into a zone maintained under pressure within the extruder.

The amount of the graft-modified polyolefin elastomer required to effectively serve as a compatibilizer will, of course, vary with the ratio of the polypropylene and the thermoplastic polyester, the chemical and physical characteristics of the polyolefin elastomer, the unsaturated organic compound containing a carboxyl group (and the extent of grafting), and similar factors. Preferably, the graft-modified polyolefin elastomer is present in an amount of at least 0.1 percent by weight, preferably at least

0.5 percent by weight, more preferably at least 1.0 percent by weight, more preferably at least 2.0 percent by weight, and most preferably at least 5.0 percent by weight based on the weight of the polymer blend composition. Generally, the graft-modified polyolefin elastomer is present in an amount less than or equal to 50 percent by weight, preferably less than or equal to 40 percent by weight, more preferably less than or equal to 30 percent by weight, more preferably less than or equal to 20 percent by weight, and most preferably less than or equal to 15 percent by weight based on the weight of the polymer blend composition.

Optionally, the polymer blend composition comprises (d) an impact modifier. Preferable impact modifiers are rubber materials having Tg's less than 0°C, preferably less than -10°C, more preferably less than -20°C and most preferably less than -30°C. Suitable rubbers include the well known homopolymers and copolymers of conjugated dienes, particularly butadiene; as well as other rubbery polymers such as acrylate rubbers, particularly homopolymers and copolymers of alkyl acrylates having from 4 to 6 carbons in the alkyl group; or polyolefin elastomers as discussed hereinabove, particularly copolymers of ethylene, propylene and optionally a nonconjugated diene. If the impact modifier component (d) is a polyolefin elastomer, it may be the same as or different from the polyolefin elastomer selected for graft modification as component (c). In addition, mixtures of the foregoing rubbery polymers may be employed if desired.

A preferred rubber is a homopolymer of butadiene and copolymer thereof with up to 30 percent by weight styrene. Such copolymers may be random or block copolymers and in addition may be hydrogenated to remove residual unsaturation. Preferably, a vinyl aromatic and conjugated diene block copolymer formed from styrene and butadiene or styrene and isoprene is used. When the styrene and butadiene copolymer is hydrogenated, it is frequently represented as styrene and (ethylene and butylene) copolymer in the di-block form, or as styrene and (ethylene and butylene) and styrene copolymer in the tri-block form. When the styrene and isoprene copolymer is hydrogenated, it is frequently represented as styrene and (ethylene and propylene) copolymer in the di-block form, or as styrene and (ethylene and propylene) and styrene copolymer in the tri-block form. Vinyl aromatic and diene block copolymers such as are described above are discussed in greater detail in Holden, US-A-3,265,766, Haelele, US-A-3,333,024, Wald, US-A-3,595,942, and Witsiepe, US-A-3,651,014, and many are available commercially as the various KRATON™ elastomers from Shell Chemical Company.

Preferably, the impact modifier is a grafted homopolymer or copolymer of

butadiene which is grafted with a polymer of styrene and methyl methacrylate. Some of the preferred rubber-containing materials of this type are the known MBS-type core/shell grafted copolymers having a  $T_g$  less than  $0^{\circ}\text{C}$  and a rubber content greater than 40 percent, typically greater than 50 percent. They are generally obtained by graft polymerizing styrene and methylmethacrylate and/or equivalent monomers in the presence of a conjugated diene polymer rubber core, preferably a butadiene homo- or co-polymer. The grafting monomers may be added to the reaction mixture simultaneously or in sequence, and, when added in sequence, layers, shells or wart-like appendages can be built up around the substrate latex, or core. The monomers can be added in various ratios to each other.

Other impact modifiers useful in the compositions of this invention are those based generally on a long-chain, hydrocarbon backbone, which may be prepared predominantly from various mono- or dialkenyl monomers and may be grafted with one or more styrenic monomers. Representative examples of a few olefinic elastomers which illustrate the variation in the known substances which would suffice for such purpose are as follows: butyl rubber; chlorinated polyethylene rubber; chlorosulfonated polyethylene rubber; an olefin polymer or copolymer such as ethylene/propylene copolymer, ethylene/styrene copolymer or ethylene/propylene/diene copolymer, which may be grafted with one or more styrenic monomers; neoprene rubber; nitrile rubber; polybutadiene and polyisoprene.

If used, the impact modifier is preferably present in an amount of at least 1 percent by weight, preferably at least 2 percent by weight, more preferably at least 5 percent by weight, more preferably at least 10 percent by weight, and most preferably at least 15 percent by weight based on the weight of the polymer blend composition. Generally, the impact modifier is present in an amount less than or equal to 50 percent by weight, preferably less than or equal to 40 percent by weight, more preferably less than or equal to 30 percent by weight, more preferably less than or equal to 25 percent by weight, and most preferably less than or equal to 20 percent by weight based on the weight of the polymer blend composition.

The claimed polymer blend compositions may also optionally contain a component (e) which is one or more additives that are commonly used in polymer blend compositions of this type. Preferred additives of this type include, but are not limited to: fillers, reinforcements, ignition resistant additives, stabilizers, colorants, antioxidants, antistats, flow enhancers, mold releases, nucleating agents, etc. Preferred examples of additives are fillers, such as, but not limited to talc, clay, wollastonite, mica, glass or a mixture thereof. Additionally, ignition resistance

additives, such as, but not limited to halogenated hydrocarbons, halogenated carbonate oligomers, halogenated diglycidyl ethers, organophosphorous compounds, fluorinated olefins, antimony oxide and metal salts of aromatic sulfur, or a mixture thereof may be used. Further, compounds which stabilize polymer blend  
5 compositions against degradation caused by, but not limited to heat, light, and oxygen, or a mixture thereof may be used.

If used, such additives may be present in an amount from at least 0.01 percent by weight, preferably at least 0.1 percent by weight, more preferably at least 1 percent by weight, more preferably at least 2 percent by weight, and most preferably at least 5  
10 percent by weight based on the weight of the polymer blend composition. Generally, the additive is present in an amount less than or equal to 25 percent by weight, preferably less than or equal to 20 percent by weight, more preferably less than or equal to 15 percent by weight, more preferably less than or equal to 12 percent by weight, and most preferably less than or equal to 10 percent by weight based on the  
15 weight of the polymer blend composition.

Preparation of the polymer blend compositions of this invention can be accomplished by any suitable mixing means known in the art, including dry blending the individual components and subsequently melt mixing, either directly in the extruder used to make the finished article (for example, the automotive part), or pre-  
20 mixing in a separate extruder (for example, a Banbury mixer). Dry blends of the compositions can also be directly injection molded without pre-melt mixing.

When softened or melted by the application of heat, the polymer blend compositions of this invention can be formed or molded using conventional techniques such as compression molding, injection molding, gas assisted injection  
25 molding, calendering, vacuum forming, thermoforming, extrusion and/or blow molding, alone or in combination. The polymer blend compositions can also be formed, spun, or drawn into films, fibers, multi-layer laminates or extruded sheets, or can be compounded with one or more organic or inorganic substances, on any machine suitable for such purpose. Some of the fabricated articles include automotive bumper  
30 beams, bumper facia, pillars, and internal trims; in filaments such as yarns and fibers; in wire and cable jackets and tubings; and in electrical and electrical equipment device housing and covers; as well as other household and personal articles, including, for example, freezer containers.

To illustrate the practice of this invention, examples of preferred embodiments  
35 are set forth below. However, these examples do not in any manner restrict the scope of this invention.

The compositions of Examples 1 to 4 were prepared by mixing the dry components in a tumble mixer, and then feeding the dry-blended formulation to a 30 mm Werner and Pfleider extruder. The following were the compounding conditions on the Werner and Pfleider extruder: Barrel temperature profile: 190, 240, 270, 270, and 270 °C; RPM: 350; Torque: 33 percent. The extrudate is cooled in the form of strands and comminuted as pellets. The pellets are dried in an air draft oven for 3 hours at 120 °C, and then are used to prepare test specimens on a 70 ton Arburg injection molding machine, having the following molding conditions: Barrel temperatures of 258, 257, 254, and 254 °C; Mold temperature was ambient temperature; Injection pressure: 30 bar; Holding pressure: 25 bar; Back pressure: 0 bar; Screw speed: 3.1; Injection speed: 3.1; Injection time: 3 seconds; Holding time: 19 seconds; and Cooling time: 20 seconds.

The formulation content and properties of Examples 1 to 4 are given in Table 1 below in percent by weight of the total composition. In Tables 1:

PP is isotactic polypropylene homopolymer commercially available as H702-20 from Dow having a 20 MI;

PET is polyethylene terephthalate which is commercially available as LIGHTER™ C88 from INCA having an intrinsic viscosity of 0.77 deciliter per gram (dl/g) measured according to INCA analytical method 1/MA/1/002 and a density of 1.39 g/cm<sup>3</sup>;

MAH-g-EPDM is maleic anhydride grafted on to ethylene propylene rubber with some diene monomer which is commercially available as ROYALTUFF™ 465 from Uniroyal Chemical Company;

MAH-g-SLEP is a substantially linear ethylene polymer commercially available as ENGAGE™ SM 8180 from DuPont/Dow Elastomers grafted with 1 weight percent maleic anhydride and having a MI after grafting of 0.56.

The following tests were run on Examples 1 to 4 and the results of these test are shown in Table 1:

Impact resistance as measured by the Notched Izod test (Izod) was determined according to ASTM D 256-90-B at room temperature. Specimens were cut from rectangular DTUL bars and measured 3.18 millimeter (mm) in thickness and 50.8 mm in length. The specimens were notched with a TMI 22-05 notcher to give a 0.254 mm (10 mil) radius notch. A 22 kilogram pendulum was used.

Impact resistance as measured by instrumented impact (Dart Impact) was determined according to ASTM D 3763 using a General Research Corp. Dynatup

8250 instrumented impact tester with a 45.4 kg weight. Test results were determined at room temperature on a 64 mm by 3.18 mm thick disk.

Tensile property testing was done in accordance with ASTM D 638. Tensile Type 1 test specimens were conditioned at 23 °C and 50 percent relative humidity 24 hours prior to testing. Testing was performed using an INSTRON 1125 mechanical tester. Testing was performed at room temperature.

Flexural properties were determined in accordance with ASTM D 790. Testing was performed using an INSTRON mechanical tester. Flexural property test specimens were conditioned at 23 °C and 50 percent relative humidity 24 hours prior to testing. Testing was performed at room temperature.

Deflection temperature under load (DTUL) was determined on a Ceast HDT 300 Vicat machine in accordance with ASTM D 648-82 where test specimens were unannealed and tested under applied pressures of 0.46 MPa and 1.82 MPa.

MFR was determined according to ASTM D 1238 on a Tinius Olsen plastometer, at conditions of 230 °C and an applied load of 3.8 kg.

Dynamic Mechanical Spectroscopy (DMS) was determined on a Rheometrix RMS 800 dynamic mechanical spectroscopy using a 50.8 mm length specimen cut from the DTUL specimen. Testing was done from -150 °C to the blend's melting point at a step of 5 °C per step and a frequency of 1 Hertz (Hz).

Solvent testing was conducted on tensile bars placed on a jig and aligned horizontally by adjusting a micrometer. Once aligned, the micrometer is adjusted so that the specimen is bent to a deflection that will yield a strain of 0.05 and 1.25 percent. The formula used to convert deflection to strain is:

$$\delta = \frac{I^2 \varepsilon}{100Lt}$$

where  $I$  is the span length between mounting pins,  $\varepsilon$  is the desired strains,  $L$  is the length of the specimen and  $t$  is the thickness, and  $\delta$  is the deflection. The specimens were immersed in a mixture of 60/40 iso-octane/toluene for 3, 5, and 10 minutes. For each immersion time, the specimens were taken out and observed for cracks followed by a tensile test. Elongation at break was recorded as determining the toughness after solvent exposure.



Table 1.

Example	1	2	3	4
COMPOSITION				
PP	45	45	20	70
PET	45	45	70	20
MAH-g-SLEP	-	10	10	10
MAH-g-EPDM	10	-	-	-
PROPERTIES				
Notched Izod, J/m	42	64	144	90.8
Dart Impact, J, Total	1.6	18.6	3.6	29.2
Tensile Properties				
Yield, MPa	4.2	3.1	34	21
Modulus, MPa	1600	810	-	-
Elongation, percent	9	29	56	77
Flexural Modulus, MPa	1660	1180	-	-
DTUL, °C				
@ 0.45 MPa	75	62	-	-
@ 1.82 MPa	60	44	57	47
Melt Flow Rate, @ 230 °C/3.8 kg, g/10 min	0.4	2.1	-	-
DMS Storage Modulus (G') @ 150°C, MPa	52	25	-	-
Solvent Crack, 10 min.	none	none	-	-
Solvent Elongation at break, percent	7.9	40	-	-

## CLAIMS:

1. A polymer blend composition comprising:
  - (a) a polypropylene in an amount from 95 to 5 percent by weight,
  - (b) a thermoplastic polyester in an amount from 5 to 95 percent by weight,
  - 5 (c) a polyolefin elastomer grafted with at least 0.01 weight percent of an unsaturated organic compound containing, prior to grafting, at least one site of ethylenic unsaturation and at least one carbonyl group, in an amount from 0.1 to 50 percent by weight,
  - and
  - 10 (d) an impact modifier in an amount from 0 to 50 percent by weight,wherein percent by weight is based on the weight of the polymer blend composition.
2. The polymer blend composition of Claim 1 wherein the unsaturated organic compound is maleic anhydride.
3. The polypropylene blend composition of Claim 1 wherein the polyolefin  
15 elastomer is a substantially linear ethylene polymer or linear ethylene polymer characterized as having:
  - (a) a density of less than 0.93 g/cm<sup>3</sup>,
  - (b) a molecular weight distribution,  $M_w/M_n$ , of less than 3.0,
  - and
  - 20 (c) a Composition Distribution Branch Index of greater than 50 percent.
4. The polymer blend composition of Claim 3 wherein the substantially linear ethylene polymer or linear ethylene polymer is a copolymer of ethylene with a C<sub>3</sub> to C<sub>20</sub> alpha-olefin.
5. The polymer blend composition of Claim 3 wherein the substantially linear  
25 ethylene polymer or linear ethylene polymer is a copolymer of ethylene with propylene, 1-butene, 1-hexene or 1-octene.
6. The polymer blend composition of Claim 3 wherein the substantially linear ethylene polymer or linear ethylene polymer is a copolymer of ethylene and 1-octene.
7. The polymer blend composition of Claim 1 wherein the polyolefin elastomer is  
30 an ethylene, propylene, and non-conjugated diene terpolymer.
8. The polymer blend composition of Claim 1 wherein the polypropylene is isotactic.
9. The polymer blend composition of Claim 1 wherein the polyester is polyethylene terephthalate.
- 35 10. The polymer blend composition of Claim 1 further comprising a filler.

11. The polymer blend composition of Claim 10 wherein the filler is talc, wollastonite, clay, mica, glass or a mixture thereof.

12. The polymer blend composition of Claim 10 wherein the filler is talc.

13. The polymer blend composition of Claim 1 further comprising one or more  
5 ignition resistance additives selected from halogenated hydrocarbons, halogenated carbonate oligomers, halogenated diglycidyl ethers, organophosphorous compounds, fluorinated olefins, antimony oxide and metal salts of aromatic sulfur compounds.

14. The polymer blend composition of Claim 1 wherein the polypropylene is an isotactic polypropylene, the thermoplastic polyester is polyethylene terephthalate, the  
10 polyolefin elastomer is a substantially linear ethylene polymer which is a copolymer of ethylene and 1-octene and the unsaturated organic compound is maleic anhydride.

15. A method for preparing a polymer blend composition comprising the step of combining:

- (a) a polypropylene in an amount from 95 to 5 percent by weight,
- 15 (b) a thermoplastic polyester in an amount from 5 to 95 percent by weight,
- (c) a polyolefin elastomer grafted with at least 0.01 weight percent of an unsaturated organic compound containing, prior to grafting, at least one site of ethylenic unsaturation and at least one carbonyl group, in an amount from 0.1 to 50 percent by weight,
- 20 and
- (d) an impact modifier in an amount from 0 to 50 percent by weight,

wherein percent by weight is based on the weight of the polymer blend composition.

16. The method according to Claim 15 wherein the polypropylene is an isotactic polypropylene, the thermoplastic polyester is polyethylene terephthalate, the  
25 polyolefin elastomer is a substantially linear ethylene polymer which is a copolymer of ethylene and 1-octene, and the unsaturated organic compound is maleic anhydride.

17. The composition of Claim 1 in the form of a molded or extruded article.

# INTERNATIONAL SEARCH REPORT

International Application No.

PCT/US 00/01725

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08L23/10 C08L67/00 //(C08L23/10,51:06,67:00)

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 866 081 A (KANEKAFUCHI CHEMICAL IND) 23 September 1998 (1998-09-23)	1-13, 15, 17
Y	example 18	14, 16
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Y	example 19	14, 16
Y	US 5 346 963 A (ROWLAND MICHAEL E ET AL) 13 September 1994 (1994-09-13) cited in the application column 4, line 57 -column 5, line 41	14, 16
Y	US 5 280 066 A (TEKKANAT BORA ET AL) 18 January 1994 (1994-01-18) cited in the application table 3	14, 16

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

2 May 2000

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Schmidt, H

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 00/01725

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DERWENT-ACC-NO: 2000-601883

DERWENT-WEEK: 200362

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**TITLE:** Polypropylene and polyester blend useful in the form of molded articles e.g. pillars, sheets comprises a graft-modified polyolefin elastomer

**INVENTOR:** PHAM H T; WU S**PATENT-ASSIGNEE:** DOW CHEM CO[DOWC]**PRIORITY-DATA:** 1999US-262478 (March 4, 1999)**PATENT-FAMILY:**

PUB-NO	PUB-DATE	LANGUAGE
WO 0052094 A1	September 8, 2000	EN
AU 200026261 A	September 21, 2000	EN
EP 1165685 A1	January 2, 2002	EN
BR 200010270 A	January 22, 2002	PT
KR 2001108304 A	December 7, 2001	KO
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**DESIGNATED-STATES:** AE AL AM AT AU AZ BA BB BG BR BY CA CH CN  
 CR CU CZ DE DK DM EE ES FI GB GD GE GH GM  
 HR HU ID IL IN IS JP KE KG KP KR KZ LC LK LR LS  
 LT LU LV MA MD MG MK MN MW MX NO NZ PL PT  
 RO RU SD SE SG SI SK SL TJ TM TR TT T Z UA UG  
 UZ VN YU ZA ZW AT BE CH CY DE DK EA ES FI FR  
 GB GH GM GR IE IT KE LS LU MC MW NL OA PT SD  
 SE SL SZ TZ UG ZW AL AT BE CH CY DE DK ES FI  
 FR GB GR IE IT LI LT LU LV MC MK NL PT RO SE SI

**APPLICATION-DATA:**



PUB-NO	APPL-DESCRIPTOR	APPL-NO	APPL-DATE
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KR2001108304A	N/A	2001KR- 711207	September 3, 2001
MX2001008924A1	Based on	2001MX- 008924	September 4, 2001

## INT-CL-CURRENT:

TYPE	IPC DATE
CIPS	C08L23/10 20060101
CIPS	C08L67/02 20060101

ABSTRACTED-PUB-NO: WO 0052094 A1

## BASIC-ABSTRACT:



**NOVELTY** - A polymer blend composition comprises (wt.%): (a) a polypropylene (5 - 95); (b) a thermoplastic polyester (95 - 5); (c) a polyolefin elastomer (0.1 - 50) grafted with an unsaturated organic compound (at least 0.01) containing, before grafting, at least one site of ethylenic unsaturation and at least one carbonyl group; and (d) an impact modifier (0 - 50).

**DESCRIPTION** - An INDEPENDENT CLAIM is included for preparing the composition by combining the polypropylene, the thermoplastic polyester, the polyolefin elastomer and the impact modifier.

**USE** - In the form of molded or extruded articles (claimed), such as automotive bumper beams, bumper facia, pillars and internal trims; films; fibers; multi-layer laminates; extruded sheets; in filaments such as yarns and fibers; in wire and cable jackets and tubings; and in electrical and electrical equipment device housing and covers; as well as other household and personal articles e.g. freezer containers.

**ADVANTAGE** - The composition has improved processability and when molded, having improved strength. The composition possess a good balance of good processability, good thermal and physical properties, good solvent resistance and especially impact resistance.

**EQUIVALENT-ABSTRACTS:**

## INORGANIC CHEMISTRY

**Preferred Additives:** The filler is talc, wallstonite, clay, mica and/or glass (preferably talc). The ignition resistance additive is selected from halogenated hydrocarbon, halogenated carbonate oligomer, halogenated diglycidyl ether, organophosphorous compound, fluorinated olefin, antimony oxide and metal salt of aromatic sulfur compound.

## POLYMERS

**Preferred Composition:** The polyolefin elastomer is a linear ethylene polymer having a density of less than 0.93 g/cm<sup>3</sup>, a molecular weight distribution (Mw/Mn) of less than 3 and a Composition Distribution Branch Index of greater than 50%.

**Preferred Components:** The linear ethylene polymer is a copolymer of ethylene with propylene, 1-butene, 1-hexene or 1-octene (preferably 1-octene). The polyolefin elastomer is an ethylene, propylene and non-



conjugated diene terpolymer. The polypropylene is isotactic and the polyester is a polyethylene terephthalate. The composition further comprises a filler and at least one ignition resistance additive.

## ORGANIC CHEMISTRY

**Preferred Additives:** The ignition resistance additive is selected from halogenated hydrocarbon, halogenated carbonate oligomer, halogenated diglycidyl ether, organophosphorous compound, fluorinated olefin and metal salt of aromatic sulfur compound.

## SPECIFIC COMPOUNDS

Maleic anhydride is specifically claimed as the unsaturated organic compound.

The blend composition was prepared by mixing the following dry components (wt.%) in a tumble mixer: H702-20 (isotactic polypropylene homopolymer (PP)) (20), LIGHTER C88 (RTM; polyethylene terephthalate (PET)) (70), ENGAGE SM 8180 (RTM; linear ethylene polymer (MAH-g-SLEP)) (10). Then the composition was fed to an extruder, at barrel temperature profile of 190, 240, 270, 270 and 270degreesC. The extrudate was cooled in the form of strands and communicated as pellets. The pellets were dried in an air draft oven for 3 hours at 120degreesC and then were used to prepare test specimens on a 70 ton Arburg injection molding machine which had the barrel temperature of 258, 257, 254 and 254degreesC. The injection pressure was 30 bar, had injection time of 3 seconds and cooling time of 20 seconds. The prepared article had impact resistance of 144 J/m, elongation of 56% and deflection temperature under load (DTUL) of 57degreesC at 1.82 Mpa.

**TITLE-TERMS:** POLYPROPYLENE POLYESTER BLEND USEFUL FORM  
ARTICLE PILLAR SHEET COMPRISE GRAFT MODIFIED  
POLYOLEFIN ELASTOMER

**DERWENT-CLASS:** A17 A23 A85 A95 F01

**CPI-CODES:** A04-G03B; A05-E01A2; A10-E03; F01-D04; F01-D05;



**ENHANCED-POLYMER-INDEXING:** Polymer Index [1.1] 018 ; H0022 H0011;  
 G0044 G0033 G0022 D01 D02 D12 D10  
 D51 D53 D58 D82 R00326 1013; G0044  
 G0033 G0022 D01 D02 D12 D10 D51  
 D53 D58 D83 R00964 1145; S9999  
 S1434; S9999 S1581; S9999 S1285\*R;  
 S9999 S1070\*R; S9999 S1547 S1536;  
 P1150; P1285;

Polymer Index [1.2] 018 ; H0022 H0011;  
 G0044 G0033 G0022 D01 D02 D12 D10  
 D51 D53 D58 D82 R00326 1013; G0055  
 G0044 G0033 G0022 D01 D02 D12 D10  
 D51 D53 D58 D84 R00805 11179; S9999  
 S1434; S9999 S1581; S9999 S1285\*R;  
 S9999 S1070\*R; S9999 S1547 S1536;  
 P1150; P1263;

Polymer Index [1.3] 018 ; H0022 H0011;  
 G0044 G0033 G0022 D01 D02 D12 D10  
 D51 D53 D58 D82 R00326 1013; G0044  
 G0033 G0022 D01 D02 D12 D10 D51  
 D53 D58 D86 R02043 785; S9999  
 S1434; S9999 S1581; S9999 S1285\*R;  
 S9999 S1070\*R; S9999 S1547 S1536;  
 P1150;

Polymer Index [1.4] 018 ; H0022 H0011;  
 G0044 G0033 G0022 D01 D02 D12 D10  
 D51 D53 D58 D82 R00326 1013; G0044  
 G0033 G0022 D01 D02 D12 D10 D51  
 D53 D58 D88 R00936 251; S9999  
 S1434; S9999 S1581; S9999 S1285\*R;  
 S9999 S1070\*R; S9999 S1547 S1536;  
 P1150;

Polymer Index [1.5] 018 ; K9745\*R;  
 K9449; Q9999 Q9234 Q9212; Q9999  
 Q9289 Q9212; Q9999 Q7818\*R; Q9999  
 Q7330\*R; Q9999 Q7681\*R; B9999  
 B3623 B3554; B9999 B4091\*R B3838  
 B3747; B9999 B4626 B4568; B9999  
 B4159 B4091 B3838 B3747; N9999  
 N5970\*R; N9999 N6597 N6586; N9999



N6780\*R N6655; B9999 B3907 B3838  
B3747; B9999 B5594 B5572; ND04;

Polymer Index [1.6] 018 ; G3190 D00  
F80 O\* 6A Mg 2A Si 4A R01541 108296  
147554; D00 F80 O\* 6A Al 3A Si 4A  
R01949 129788; A999 A237;

Polymer Index [1.7] 018 ; G3418 D00  
F80 O\* 6A Ca 2A Si 4A; A999 A237;

Polymer Index [1.8] 018 ; G3010 D00  
F80 Al 3A Si 4A O\* 6A; A999 A237;

Polymer Index [1.9] 018 ; G2880 D00 Si  
4A; A999 A237;

Polymer Index [1.10] 018 ; D01 D12 D10  
D51\*R D69 7A\*R; A999 A248\*R;

Polymer Index [1.11] 018 ; D01 P\* 5A;  
A999 A248\*R;

Polymer Index [1.12] 018 ; G2482\*R  
D00 F20 O\* 6A Sb 5A; A999 A248\*R;

Polymer Index [1.13] 018 ; D01 D18\*R  
S\* 6A; A999 A248\*R;

Polymer Index [2.1] 018 ; G0817\*R D01  
D51 D54; G0044 G0033 G0022 D01 D02  
D12 D10 D51 D53 D58 D82 R00326  
1013; G0044 G0033 G0022 D01 D02  
D12 D10 D51 D53 D58 D83 R00964  
1145; H0033 H0011; H0135 H0124;  
S9999 S1434; S9999 S1581; M9999  
M2299; M9999 M2368; S9999 S1285\*R;  
S9999 S1070\*R; S9999 S1547 S1536;  
P1150;

Polymer Index [2.2] 018 ; K9745\*R;  
K9449; Q9999 Q9234 Q9212; Q9999



Q9289 Q9212; Q9999 Q7818\*R; Q9999  
 Q7330\*R; Q9999 Q7681\*R; B9999  
 B3623 B3554; B9999 B4091\*R B3838  
 B3747; B9999 B4626 B4568; B9999  
 B4159 B4091 B3838 B3747; N9999  
 N5970\*R; N9999 N6597 N6586; N9999  
 N6780\*R N6655; B9999 B3907 B3838  
 B3747; B9999 B5594 B5572; ND04;

Polymer Index [2.3] 018 ; G0760 G0022  
 D01 D23 D22 D31 D42 D51 D53 D59  
 D65 D75 D84 F39 E00 E01 R00843 790;  
 H0226;

Polymer Index [2.4] 018 ; G3190 D00  
 F80 O\* 6A Mg 2A Si 4A R01541 108296  
 147554; D00 F80 O\* 6A Al 3A Si 4A  
 R01949 129788; A999 A237;

Polymer Index [2.5] 018 ; G3418 D00  
 F80 O\* 6A Ca 2A Si 4A; A999 A237;

Polymer Index [2.6] 018 ; G3010 D00  
 F80 Al 3A Si 4A O\* 6A; A999 A237;

Polymer Index [2.7] 018 ; G2880 D00 Si  
 4A; A999 A237;

Polymer Index [2.8] 018 ; D01 D12 D10  
 D51\*R D69 7A\*R; A999 A248\*R;

Polymer Index [2.9] 018 ; D01 P\* 5A;  
 A999 A248\*R;

Polymer Index [2.10] 018 ; G2482\*R  
 D00 F20 O\* 6A Sb 5A; A999 A248\*R;

Polymer Index [2.11] 018 ; D01 D18\*R  
 S\* 6A; A999 A248\*R;

Polymer Index [3.1] 018 ; P0884 P1978  
 P0839 H0293 F41 D01 D11 D10 D19



D18 D31 D50 D63 D90 E21 E00; S9999  
S1434; S9999 S1581; S9999 S1285\*R;  
S9999 S1070\*R; S9999 S1547 S1536;

Polymer Index [3.2] 018 ; K9745\*R;  
K9449; Q9999 Q9234 Q9212; Q9999  
Q9289 Q9212; Q9999 Q7818\*R; Q9999  
Q7330\*R; Q9999 Q7681\*R; B9999  
B3623 B3554; B9999 B4091\*R B3838  
B3747; B9999 B4626 B4568; B9999  
B4159 B4091 B3838 B3747; N9999  
N5970\*R; N9999 N6597 N6586; N9999  
N6780\*R N6655; B9999 B3907 B3838  
B3747; B9999 B5594 B5572; ND04;

Polymer Index [3.3] 018 ; G3190 D00  
F80 O\* 6A Mg 2A Si 4A R01541 108296  
147554; D00 F80 O\* 6A Al 3A Si 4A  
R01949 129788; A999 A237;

Polymer Index [3.4] 018 ; G3418 D00  
F80 O\* 6A Ca 2A Si 4A; A999 A237;

Polymer Index [3.5] 018 ; G3010 D00  
F80 Al 3A Si 4A O\* 6A; A999 A237;

Polymer Index [3.6] 018 ; G2880 D00 Si  
4A; A999 A237;

Polymer Index [3.7] 018 ; D01 D12 D10  
D51\*R D69 7A\*R; A999 A248\*R;

Polymer Index [3.8] 018 ; D01 P\* 5A;  
A999 A248\*R;

Polymer Index [3.9] 018 ; G2482\*R D00  
F20 O\* 6A Sb 5A; A999 A248\*R;

Polymer Index [3.10] 018 ; D01 D18\*R  
S\* 6A; A999 A248\*R;

Polymer Index [4.1] 018 ; G0044 G0033



G0022 D01 D02 D12 D10 D51 D53 D58  
D83 R00964 1145; H0000; S9999  
S1434; S9999 S1581; S9999 S1285\*R;  
S9999 S1070\*R; S9999 S1547 S1536;  
P1150; P1343;

Polymer Index [4.2] 018 ; B9999 B4955  
B4944 B4922 B4740;

Polymer Index [4.3] 018 ; K9745\*R;  
K9449; Q9999 Q9234 Q9212; Q9999  
Q9289 Q9212; Q9999 Q7818\*R; Q9999  
Q7330\*R; Q9999 Q7681\*R; B9999  
B3623 B3554; B9999 B4091\*R B3838  
B3747; B9999 B4626 B4568; B9999  
B4159 B4091 B3838 B3747; N9999  
N5970\*R; N9999 N6597 N6586; N9999  
N6780\*R N6655; B9999 B3907 B3838  
B3747; B9999 B5594 B5572; ND04;

Polymer Index [4.4] 018 ; G3190 D00  
F80 O\* 6A Mg 2A Si 4A R01541 108296  
147554; D00 F80 O\* 6A Al 3A Si 4A  
R01949 129788; A999 A237;

Polymer Index [4.5] 018 ; G3418 D00  
F80 O\* 6A Ca 2A Si 4A; A999 A237;

Polymer Index [4.6] 018 ; G3010 D00  
F80 Al 3A Si 4A O\* 6A; A999 A237;

Polymer Index [4.7] 018 ; G2880 D00 Si  
4A; A999 A237;

Polymer Index [4.8] 018 ; D01 D12 D10  
D51\*R D69 7A\*R; A999 A248\*R;

Polymer Index [4.9] 018 ; D01 P\* 5A;  
A999 A248\*R;

Polymer Index [4.10] 018 ; G2482\*R  
D00 F20 O\* 6A Sb 5A; A999 A248\*R;



Polymer Index [4.11] 018 ; D01 D18\*R  
S\* 6A; A999 A248\*R;

Polymer Index [5.1] 018 ; D01 D11 D10  
D23 D22 D73 D42 D50 D69 F34 7A\*R;  
P0464\*R D01 D22 D42 F47; A999  
A248\*R; A999 A782;

Polymer Index [6.1] 018 ; 7A\*R; P0862  
P0839 F41 F44 D01 D63; H0237\*R;  
A999 A248\*R; A999 A782;

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